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## Aluminum break-point contacts

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(Received 9 December 1996)

*Ab initio* molecular dynamics is used to study the contribution of a single Al atom to an aluminum break-point contact during the final stages of breaking and the initial stages of the formation of such a contact. A hysteresis effect is found in excellent agreement with experiment and the form of the conductance steps before breaking of the contact is discussed. [S0163-1829(97)12116-6]

### I. INTRODUCTION

In the last couple of years measurements on mechanically controllable break junctions (MCBJ) (Ref. 1) and scanning tunneling microscope (STM) measurements on metallic point contacts<sup>2-4</sup> have shown that for a variety of metals the conductance exhibits jumps which are more or less quantized in multiples of  $G_0 = 2e^2/h$  whenever one or more atoms stop contributing to the conductance because they are mechanically torn off the contact. Al and Pt show a more complicated and even more interesting feature: the steps in the conductance are not flat but have a positive slope as can be seen in Fig. 1. This somewhat counterintuitive result means that the conductance increases although the electrodes are pulled apart. On the theoretical side there have been some calculations using the free-electron and tight-binding approach in order to describe contact breaking and conductance.<sup>5</sup> In this paper we follow quite a different strategy: an Al point contact set up in the most simple way is treated with *ab initio* molecular dynamics in order to study the contribution of an individual atom breaking off such a point contact.

The paper is organized as follows. Section II contains an outline of the computational method. In Sec. III we present the results for the aluminum break-point contacts. In particular, we discuss the hysteresis effect and the conductivity. Finally, the paper is concluded with a summary in Sec. IV.

### II. COMPUTATIONAL METHOD

We perform self-consistent density-functional theory (DFT) (Refs. 6 and 7) calculations using the local-density approximation (LDA) for the exchange-correlation functional.<sup>8,9</sup> The atomic potentials are represented by a Kleinman-Bylander type *ab initio* pseudopotential.<sup>10</sup> The integration in  $\mathbf{k}$  space is replaced by a summation over 3, 6, and 12 special  $\mathbf{k}$  points in the irreducible part of the Brillouin zone.<sup>11</sup> We use a plane wave basis set with a kinetic energy up to 9 Ry. The atomic and electronic ground state of the system is reached using a Car-Parrinello-like scheme.<sup>12,13</sup> The Al break-point contact is modeled in a rather simple way as shown in Fig. 2. Starting from a perfect six layer Al slab we remove atoms in such a way that we end up with three complete Al layers with nine atoms per layer. On each of the (111) surfaces we keep a triangular pyramid. Just halfway between these two pyramids sits a single atom. All atoms with the exception of the middle full Al layer, i.e., the one

farthest away from the single atom, are allowed to relax. The breaking of the contact will be achieved by slowly increasing the cell length along the [111] direction. The Kohn-Sham states are occupied according to the Fermi-Dirac statistic with  $k_B T = 0.001$  eV to allow for good comparison to the experiments that were performed at low temperature. The

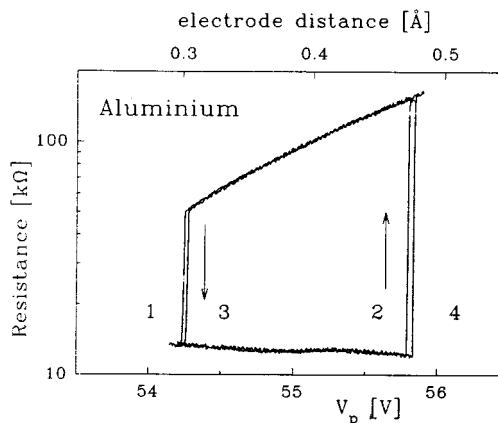
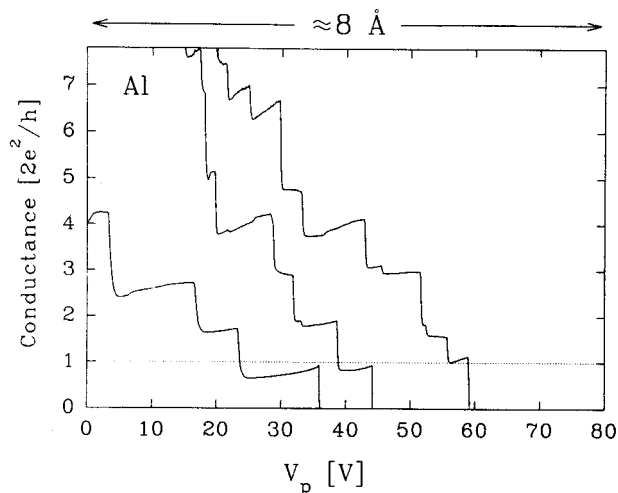


FIG. 1. The measured conductance of an Al junction at 4.2 K as a function of the piezovoltage  $V_p$ . Note that the conductance increases although the electrodes are moved apart (after Ref. 1).

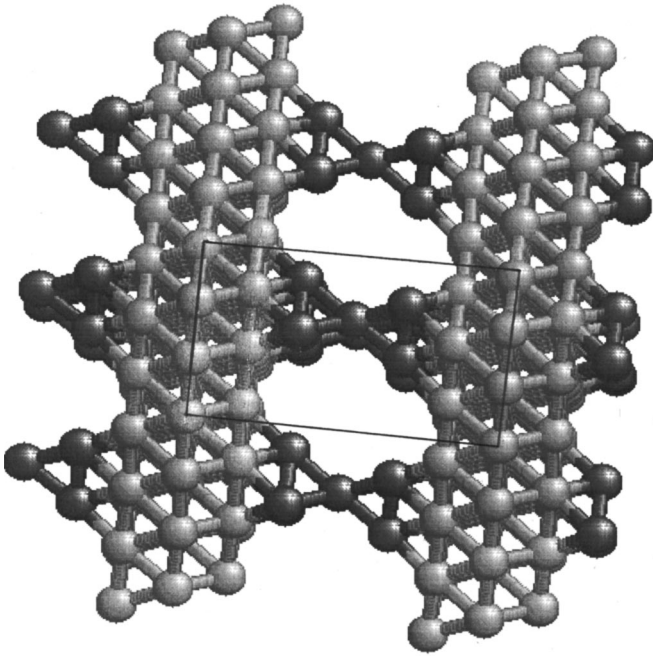


FIG. 2. Supercell used to model a break-point contact. The box indicates the unit cell of the system. The dark gray atoms form the contact and the light gray atoms are full layers of aluminum.

accuracy for the structural parameters of the system was carefully checked and uncertainties are less than  $0.02 \text{ \AA}$ .

### III. RESULTS

#### A. Atomic geometry

The entire set of calculations comprises two cycles, in each of which the contact breaking and the contact formation are simulated. Starting from all atoms in perfect fcc positions arranged as described above, we increase step by step the cell length perpendicular to the (111) surface. Due to the highly symmetrical setup this leads to symmetrical relaxations of the atoms up to a point where the induced strain becomes too big and the middle atom has to stick to one side or the other, the critical length for contact breaking. The cell is further lengthened until the total energy and potential are found to stay constant — this means that we have obtained vacuum between the surfaces. We then move the two different surfaces towards each other again by now stepwise shrinking the cell length all the way back to the perfect  $c/a$  ratio, our starting point. Note that we perform a complete calculation with relaxation of all the atomic and electronic degrees of freedom for each cell length, thereby ensuring that the system is given sufficient time to respond. In order to be able to observe possible hysteresis effects, we start each calculation with the wave functions and atomic positions of the preceding calculation. The entire cycle of calculations is performed twice to check for reproducibility.

The critical point for contact breaking is reached at an increase of the cell length of about 5.5%. The relaxations in the  $x$ - $y$  plane were not constrained by symmetry but are comparatively small for all atoms at all cell lengths — meaning  $<0.1 \text{ \AA}$ . The deviations from the perfect  $z$  coordinates can be seen to be of up to  $0.5 \text{ \AA}$ . The initial stages of cell

stretching are reflected by a nearly homogeneous stretching of the whole system. This is nicely reflected in the movement of the single atom. Figure 3 shows the  $x$ ,  $y$ , and  $z$  coordinates of the central atom as a function of the cell length. For small cell stretching it sits more or less in the middle of the cell, then starts to move to one side, and finally is bound to the underlying atoms on one surface with a binding length which is  $0.2 \text{ \AA}$  smaller than the perfect Al binding length. This result is reasonable since already a simple coordination number consideration shows that the reduced number of neighbors should immediately lead to a more closely bound adatom. On the way back — we are shrinking the cell now — we see that the central atom stays more closely bound to one surface for a longer time than on the way out. Figure 4 shows the maximal difference of the potential averaged parallel to the surfaces as a function of the cell length. This quantity is a good measure for the existence of a contact. Once the contact is broken its value should be of the order of the Al work function of  $4.25 \text{ eV}$  while it should be significantly smaller as long as the contact exists. The value of  $4.5 \text{ eV}$  that we obtain from the calculation is slightly bigger than the experimental value for Al but not unreasonable, since (a) we are performing a LDA calculation that should lead to an overestimate of the workfunction and (b) are not dealing with the ideal surface. The hysteresis effect amounts to about 1% in cell length or  $0.16 \text{ \AA}$ . It corresponds very well to the size of the effect estimated by Krans *et al.*<sup>1</sup> although we prefer not to call it electrode distance since the distance between the electrodes changes very rapidly around the critical cell length. The trajectory that the central atom follows on the way back is different from the one on the way out and is highly reproducible, as is seen from the data obtained for the second cycle. Although the lateral displacements are much smaller than those for  $z$ , they still are reproduced. The stability of the hysteresis was tested too: all atoms and electronic degrees of freedom were given random displacements of up to 5%. The hysteresis was not lifted. After the cell is shrunk to the starting length again, we find all atoms on positions very near to the starting positions; the deviations are smaller than  $0.02 \text{ \AA}$  for all atoms.

#### B. Conductance

In this subsection we would like to investigate possible reasons for the experimentally observed increase of the conductance upon increase of the electrode distance. This is a somewhat counterintuitive result since one would expect a decrease of the conductance under uniaxial stress just as for bulk aluminum. The conductance  $G$  of a system as our break-point contact is normally described within the Landauer-Büttiker formalism,<sup>14</sup>

$$G = \frac{2e^2}{h} \sum_{ab} T_{ab}(E_F), \quad (1)$$

where  $T_{ab}$  is the transmission coefficient of an incident electron in channel  $a$  on one side into channel  $b$  on the other side of the contact. Since only the electrons at the Fermi energy contribute to the conductance, we are only interested in  $T_{ab}(E_F)$ . Unfortunately, this quantity is not accessible from the type of calculation we perform and we have to therefore find another approach. Torres and Sáenz<sup>15</sup> studied the size

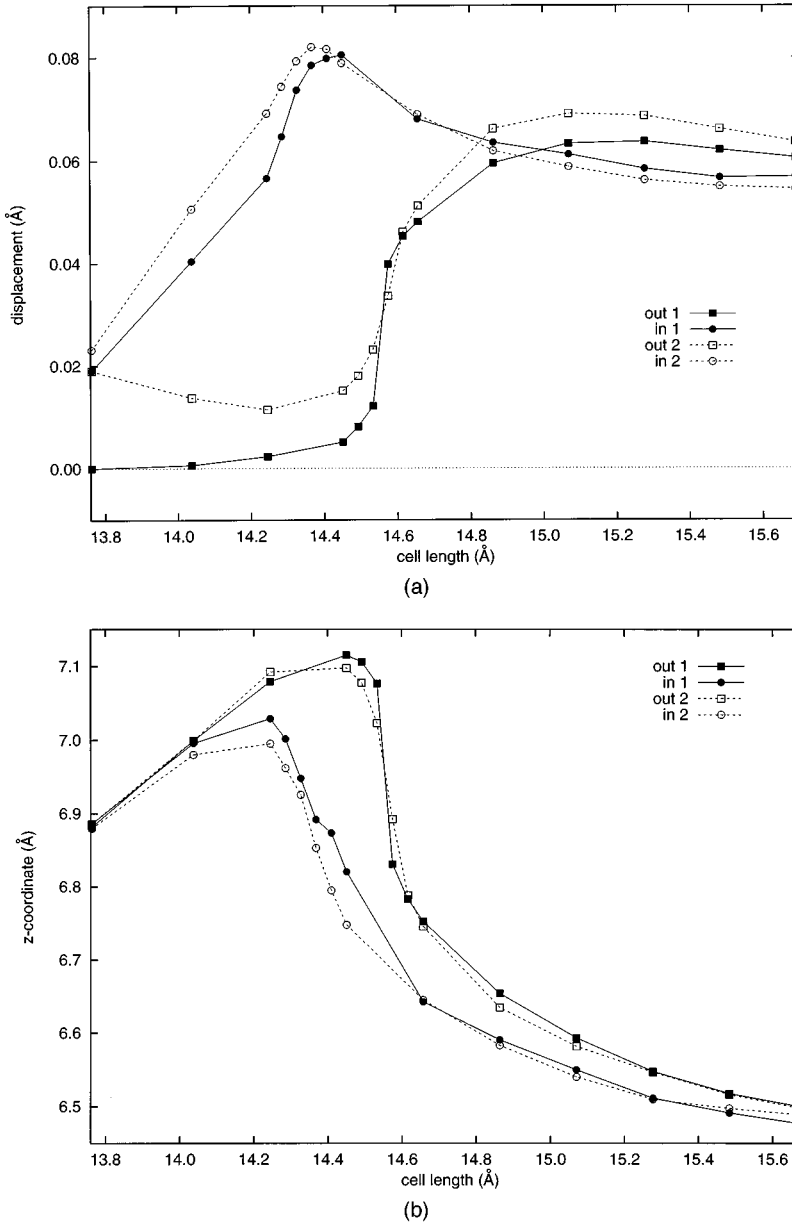


FIG. 3. Lateral displacements and  $z$  coordinates of the central atom as a function of the cell length. The two different cycles are denoted by 1 and 2. Note the high degree of reproducibility of the small but different displacements in the  $xy$  plane on the way out and in. The major contribution to the effect stems clearly from the movement parallel to  $z$ .

and shape of conductance steps in rather simply formed contacts consisting of cylindrical constrictions with variable length and radius as well as different ratios of effective atomic radius  $r_0$  to Fermi wavelength  $\lambda_F$ . For a fixed  $r_0/\lambda_F$  ratio they find very similar behavior of the conductance as a function of elongation of the contact. The result for one chosen geometry, on the other hand, shows a definitive influence of the  $r_0/\lambda_F$  ratio on the shape of the last conductance step before contact breaking. In terms of physics, this means that within the approach the contact radius renormalized with the Fermi wavelength rules not only the number of propagating modes but also the shape of the individual conductance steps crucially. So a first indication of a change in step shape might be obtained from an estimate of this quantity for the point contact system as compared to Al bulk. For both systems we can calculate the Fermi wavelength in a straightforward way:

$$\lambda_F = h \sqrt{\frac{1}{2mE_F}}. \quad (2)$$

For our first estimate we shall assume the mass,  $m$  to be the free electron mass, which should not be too bad an assumption in Al anyway. If we now further set the effective radius to be directly related to the lattice parameter  $a_l$ , we end up with

$$\frac{r_0}{\lambda_F} = \frac{a_l \sqrt{mE_F}}{2h}. \quad (3)$$

For the bulk we obtain a value of 0.389 and for the contact system we obtain a value of 0.375. Judging from the strong variation of the conductance as a function of  $r_0/\lambda_F$ , Torres and Sáenz find in the region between 0.3 and 0.4 the change in the value of  $r_0/\lambda_F$  can be taken as a first indication of the difference in the behavior of the conductance under uniaxial stress in Al bulk and break-point junctions.

#### IV. CONCLUSIONS

The calculations show the following facts.

(i) Even for the rather small system we have chosen to

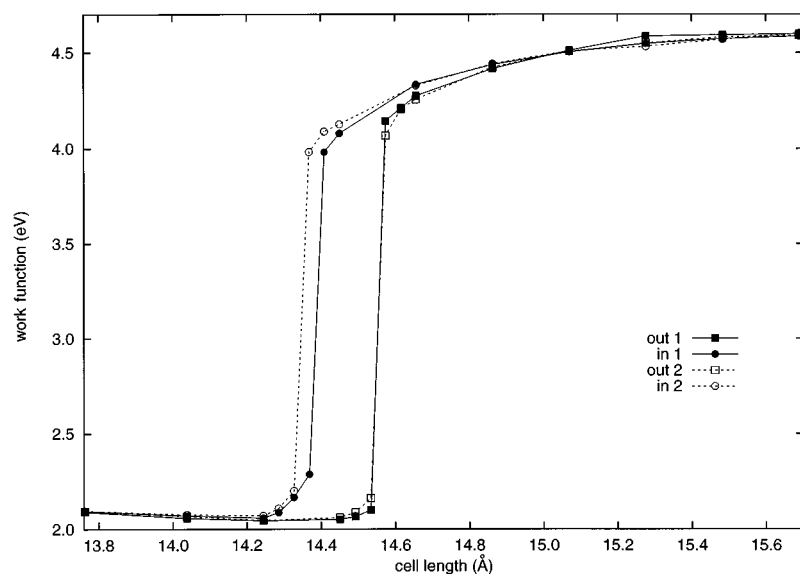


FIG. 4. Work function as calculated from the averaged electrostatic potential as a function of the cell length. The two different cycles are denoted by 1 and 2 again. The critical length for contact breaking and contact formation can be clearly seen, as can the hysteresis effect and its high degree of reproducibility.

describe, an Al break-point contact is sufficient to reproduce the experimentally observed hysteresis effect. The effect is highly reproducible and agrees well with the experimental results of Krans *et al.*<sup>1</sup> On the other hand, this means that the picture of the contact breaking being due to atomic jumps, as suggested by Krans *et al.*,<sup>1</sup> is indeed consistent with their results.

(ii) The observed increase in the conductance under elongation of the contact region seems to be linked to the change

in the ratio of contact area to Fermi wavelength as compared to Al bulk.

#### ACKNOWLEDGMENTS

It is a pleasure to thank J. M. van Ruitenbeek and J. M. Krans for fruitful discussions. This work was supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM).

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